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**[TITLE OF THE INVENTION]**

Oil-in-water Type Emulsified Composition

**[ABSTRACT]**

[Object] To provide an emulsified composition, which does not need to employ a surfactant as an emulsifying means in order to ensure high safety, sufficiently maintaining "a substantial feeling in use", and exhibiting superior stability at low temperature.

[Means for Solving the Problem] An oil-in-water type emulsified composition comprising the ingredients (A) and (B) described below:

(A) an alkyl-modified carboxyvinyl polymer and

(B) an oily ingredient in a solid state at room temperature, and having a number-average particle size of 1  $\mu$ m or less of the oily ingredient of an inner phase formed as a result of emulsification, is provided.

**[CLAIMS]**

[Claim 1] An oil-in-water type emulsified composition which comprises the ingredients (A) and (B) described below:

(A) an alkyl-modified carboxyvinyl polymer and  
(B) an oily ingredient in a solid state at room temperature, and has a number-average particle size of 1  $\mu\text{m}$  or less of the oily ingredient of an inner phase formed as a result of emulsification.

[Claim 2] The oil-in-water type emulsified composition according to Claim 1, wherein the alkyl-modified carboxyvinyl polymer is an alkyl acrylate and methacrylate copolymer.

[Claim 3] The oil-in-water type emulsified composition according to Claim 1, wherein the alkyl-modified carboxyvinyl polymer is PEMULEN TR-1 produced by BF Goodrich Co., Ltd., and/or PEMULEN TR-2 produced by the same.

[Claim 4] The oil-in-water type emulsified composition according to any one of Claims 1 to 3, wherein the alkyl-modified carboxyvinyl polymer is added in an amount of 0.01% by weight or more based on the total weight of the composition and 10% by weight or less based on the same.

[Claim 5] The oil-in-water type emulsified composition according to any one of Claims 1 to 3, wherein the alkyl-modified carboxyvinyl polymer is added in an amount of 0.05% by weight or more based on the total weight of the composition and 5% by weight or less based on the same.

[Claim 6] The oil-in-water type emulsified composition according to any one of Claims 1 to 5, wherein the oily ingredient

in a solid state at room temperature corresponds to one or more oily ingredients selected from the group consisting of higher alcohols, solid fats and oils, waxes, solid hydrocarbons, higher fatty acids, and salts of higher fatty acids.

[Claim 7] The oil-in-water type emulsified composition according to any one of Claims 1 to 5, wherein the oily ingredient in a solid state at room temperature corresponds to one or more oily ingredients of the group consisting of higher alcohols, higher fatty acids, and salts of higher fatty acids.

~~[Claim 8] The oil-in-water type emulsified composition~~  
according to Claim 6 or 7, wherein the oily ingredient in a solid state at room temperature corresponds to one or more higher alcohols selected from the group consisting of lauryl alcohol, cetyl alcohol, stearyl alcohol, behenyl alcohol, myristyl alcohol, cetostearyl alcohol, glyceryl monostearyl ether, and glyceryl monocetyl ether.

[Claim 9] The oil-in-water type emulsified composition according to Claim 6 or 7, wherein the oily ingredient in a solid state at room temperature corresponds to one or more higher fatty acids selected from the group consisting of capric acid, lauric acid, myristic acid, palmitic acid, stearic acid, arachic acid, behenic acid, lignoceric acid, cerotic acid, 2-palmitolenic acid, and erucic acid.

[Claim 10] The oil-in-water type emulsified composition according to Claim 6 or 7, wherein the oily ingredient in a solid state at room temperature corresponds to one or more higher fatty acids selected from the group consisting of capric acid,

lauric acid, myristic acid, palmitic acid, stearic acid, and arachic acid.

[Claim 11] The oil-in-water type emulsified composition according to Claim 6 or 7, wherein the oily ingredient in a solid state at room temperature corresponds to one or more salts of higher fatty acids selected from the group consisting of a caprate, a laurate, a myristate, a palmitate, a stearate, an arachate, a behenate, a lignocerate, a cerotate, a 2-palmitolenate, and an erucate.

[Claim 12] ~~The oil-in-water type emulsified composition~~ according to Claim 11, wherein the salt of a higher fatty acid corresponds to one or more salts of a higher fatty acid selected from the group consisting of a sodium salt, a potassium salt, an ammonium salt, a triethanolamine salt, a tripropanolamine salt, an aluminum salt, a magnesium salt, a zinc salt, a calcium salt, an alginate salt, and a lysine salt of a higher fatty acid.

[Claim 13] The oil-in-water type emulsified composition according to Claim 6 or 7, wherein the oily ingredient in a solid state at room temperature corresponds to one or more solid fats and oils selected from the group consisting of a cacao butter, a coconut oil, a palm oil, a palm kernel oil, a hardened oil, a hardened castor oil, a Japan wax, and a shea butter.

[Claim 14] The oil-in-water type emulsified composition according to Claim 6 or 7, wherein the oily ingredient in a solid state at room temperature corresponds to one or more waxes selected from the group consisting of beeswax, a carnauba wax,

lanolin, a reduced lanolin, a hard lanolin, a jojoba wax, and a shellac wax.

[Claim 15] The oil-in-water type emulsified composition according to Claim 6, wherein the solid hydrocarbon is vaseline and/or a microcrystalline wax.

[Claim 16] The oil-in-water type emulsified composition according to any one of Claims 1 to 15, wherein the oily ingredient in a solid state at room temperature is added in an amount of 0.5% by weight or more based on the total weight of the composition and 20% by weight or less based on the same.

[Claim 17] The oil-in-water type emulsified composition according to any one of Claims 1 to 15, wherein the oily ingredient in a solid state at room temperature is added in an amount of 1% by weight or more based on the total weight of the composition and 10% by weight or less based on the same.

[Claim 18] The oil-in-water type emulsified composition according to any one of Claims 1 to 17, obtained by emulsification by means of a nonaqueous emulsification method, a D-phase emulsification method, or a phase inversion temperature emulsification method.

[Claim 19] The oil-in-water type emulsified composition according to Claim 18, obtained by emulsification by means of a homomixer, a homodisper, a homogenizer, a colloid mill, a pebble mill, or an ultrasonic emulsifier.

[Claim 20] The oil-in-water type emulsified composition according to any one of Claims 1 to 17, obtained by emulsification by means of a high-pressure emulsifier.

[Claim 21] The oil-in-water type emulsified composition according to Claim 20, wherein the high-pressure emulsifier is a Microfluidizer, a Nanomizer, or a Mantongourin (all trade names).

[Claim 22] The oil-in-water type emulsified composition according to any one of Claims 1 to 21, which does not substantially comprise a surfactant.

#### [DETAILED DESCRIPTION OF THE INVENTION]

[0001]

[Technical Field of the Invention] The present invention belongs to the technical field relating to emulsified compositions imparting a substantial feeling in use when the composition is applied to the skin, exhibiting a superior stability at low temperature, and having high safety.

[0002]

[Prior Art] In emulsified compositions for use in cosmetics such as a cream or a milk, emulsification is essential in order to form an emulsion which is a dispersion between aqueous ingredients and oily ingredients during the steps of producing the same. As an emulsifier for adding in an emulsified composition for the purpose of emulsification, at present, various surfactants are commonly employed. For the selection criteria at the time of using a surfactant as an emulsifier, it is important to include not only exhibiting superior emulsifying effects, but also using the surfactants which can

sufficiently ensure safety for users. For these reasons, at present, surfactants exhibiting superior emulsifying effects and having superior safety are added to emulsified compositions as emulsifiers.

[0003] In addition, in order to ensure the higher safety, recently, an attempt is also made to emulsify without adding surfactants, for example, to emulsify by adding a specific alkyl-modified carboxyvinyl polymer having a high safety and exhibiting emulsifying effects to an emulsified composition, instead of adding a surfactant. However, the emulsified compositions obtained as described above tend to lack "a substantial feeling in use" which is commonly desired as features of emulsified compositions. In order to solve this problem, an attempt is made to add an oily ingredient in a solid state at room temperature during the steps of producing an emulsified composition. However, when the oily ingredient in a solid state is allowed to stand at low temperature for a long period of time, crystallization occurs, and as a result, the crystals strongly tend to deposit in the emulsified composition. For this reason, the attempt described above is not appropriate to the emulsified compositions which may be stored at low temperature such as in a cold area.

[0004]

[Problems to be Solved by the Invention] Therefore, an object to be achieved by the present invention is to provide an emulsified composition, which does not need to use a surfactant as emulsifying means in order to ensure high safety, sufficiently maintaining "a substantial feeling in use", and exhibiting superior stability at low temperature.

[0005]

[Means for Solving the Problems] The present inventors carried out diligent research in order to solve the problems described above. As a result, they discovered that in an oil-in-water type emulsified composition, the problems described above can be solved by making the number-average particle size of an oily ingredient in an inner phase 1  $\mu\text{m}$  or less, even in the case of adding a specific alkyl-modified carboxyvinyl polymer as emulsifying means and adding an oily ingredient in a solid state at room temperature in order to impart "a substantial feeling in use", in the same manner as those of the prior art described above, thus completing the present invention.

[0006] That is, the present inventors provide an oil-in-water type emulsified composition which comprises the ingredients (A) and (B) described below:

(A) an alkyl-modified carboxyvinyl polymer and

(B) an oily ingredient in a solid state at room temperature, and has a number-average particle size of 1  $\mu\text{m}$  or less of the oily ingredient of an inner phase formed as a result of emulsification.

[0007]

[Modes for Carrying Out the Invention] In the following, modes for carrying out the present invention will be explained. The alkyl-modified carboxyvinyl polymer as the ingredient (A) is different from an alkyl-modified carboxyvinyl polymer which is commonly used as a thickening agent [such as CARBOPOL 941 (produced by BF Goodrich Co., Ltd.), HIBISWAKO 105 (produced by Wako Pure Chemical Industries Ltd.), or the like], and needs to have emulsifying effects capable of emulsifying an oily ingredient in a stable state by adding the same.



[0008] As examples of the alkyl-modified carboxyvinyl polymer having these properties, mention may be made of, for example, an alkyl acrylate and methacrylate copolymer.

[0009] In addition, as the alkyl-modified carboxyvinyl polymer having these properties, although a polymer synthesized according to a commonly known method may be appropriately added as the ingredient (A) to an oil-in-water type emulsified composition of the present invention (in the following, also referred to as an "emulsified composition of the present invention"), a commercially available product can also be used. As representative examples of the commercially available products, mention may be made of PEMULEN TR-1 or PEMULEN TR-2 (both are produced by BF Goodrich Co., Ltd.).

[0010] The alkyl-modified carboxyvinyl polymer described above may be added alone to the emulsified composition of the present invention, or alternatively, may be added in combination with two or more kinds of the same.

[0011] In addition, an amount of the alkyl-modified carboxyvinyl polymer described above which is added to the emulsified composition of the present invention should be determined case by case, depending on the conditions such as formulations of the emulsified compositions of the present invention in detail. Therefore, the amount should not be particularly restricted so long as the desired effects of the present invention can be exhibited. However, it is preferable that the amount be in a range of 0.01% by weight or more and 10% by weight or less, based on the total weight of the composition. That is, if the polymer is added in an amount of below 0.01%

by weight based on the total weight of the composition, weak emulsifying effects are exhibited. On the other hand, even if the polymer is added in an amount exceeding 10% by weight based on the total weight of the composition, it is difficult to increase the emulsifying effects and emulsion stability at low temperature in proportion to increasing the amount of the polymer, and therefore, it is not preferable.

[0012] Furthermore, considering that it is desirable that a "substantial feeling in use" be imparted to the emulsified composition of the present invention as much as possible, it is particularly preferable that the alkyl-modified carboxyvinyl polymer be added in an amount of 0.05% by weight or more and 5% by weight or less, based on the total weight of the composition.

[0013] By adding the alkyl-modified carboxyvinyl polymer described above, an emulsified composition of the present invention, which does not substantially include surfactants, can be provided. That is, an emulsified composition of the present invention can be provided, in which an emulsion is formed without using surfactants commonly used as emulsifying means heretofore, that is, surfactants commonly classified as anionic surfactants, cationic surfactants, amphoteric surfactants, and nonionic surfactants.

[0014] The term "oily ingredient in a solid state at room temperature" of the ingredient (B) means an oily ingredient to be added to a composition for use in a cosmetic which is present in a solid state at room temperature, that is, at approximately 30°C. As examples of the same, mention may be made of higher alcohols, solid fats and oils, waxes, solid hydrocarbons, higher fatty acids, salts of higher fatty acids,

or the like. Among these oily ingredients, considering that a "substantial feeling in use" should be improved as much as possible, as preferable examples of the oily ingredients to be added to the emulsified composition of the present invention, mention may be made of higher alcohols, higher fatty acids, or salts of higher fatty acids (and in particular, inorganic salts).

[0015] As examples of the higher alcohols, mention may be made of, for example, a higher alcohol with a straight chain such as lauryl alcohol, cetyl alcohol, stearyl alcohol, behenyl alcohol, myristyl alcohol, or cetostearyl alcohol; a higher alcohol with a branched chain such as glyceryl monostearyl ether (batyl alcohol) or glyceryl monocetyl ether (chimyl alcohol); and the like.

[0016] As examples of the higher fatty acids, mention may be made of, for example, capric acid, lauric acid, myristic acid, palmitic acid, stearic acid, arachic acid, behenic acid, lignoceric acid, cerotic acid, 2-palmitolenic acid, erucic acid, and the like. In addition, as examples of the salts of the higher fatty acids, for example, inorganic salts such as a sodium salt, a potassium salt, or an ammonium salt; alkanol amine salts such as a triethanolamine salt, or a tripropanolamine salt; metal soaps such as an aluminum salt, a magnesium salt, a zinc salt, or a calcium salt; basic amino acid salts such as an alginate salt, or a lysine salt; or the like, of the higher fatty acids described above.

[0017] Considering that it is desirable that a "substantial feeling in use" be imparted as much as possible, it is preferable that lauric acid, myristic acid, palmitic acid, stearic acid,

arachic acid, or behenic acid, or the salts of the same be added to the emulsified composition of the present invention.

[0018] Although these salts of the fatty acids listed above also include substances which do not correspond to oily ingredients inherently, these ingredients are partially converted to a state of a fatty acid after being stored in a common emulsion for a long period of time, and therefore, solid substances may be deposited in an emulsified composition. However, as described below, even in this case, the desired effects can be exhibited in the emulsified compositions of the present invention.

[0019] In addition, as examples of the solid fats and oils, mention may be made of, for example, a cacao butter, a coconut oil, a palm oil, a palm kernel oil, a hardened oil, a hardened castor oil, a Japan wax, a shea butter, and the like. As examples of the waxes, mention may be made of, for example, beeswax, a carnauba wax, lanolin, a reduced lanolin, a hard lanolin, a jojoba wax, a shellac wax, and the like.

[0020] As examples of the solid hydrocarbons, mention may be made of, for example, vaseline, a microcrystalline wax, and the like.

[0021] These "oily ingredients in a solid state at room temperature" may be added in the emulsified composition of the present invention in an amount of 0.5% by weight or more and 20% by weight or less, based on the total weight of the composition. With the oily ingredient in an amount of below 0.5% by weight based on the total weight of the composition, effects of improving a "substantial feeling in use" cannot be exhibited. On the other

hand, even if the oily ingredient is added in an amount exceeding 20% by weight based on the total weight of the composition, the effects of improving a "substantial feeling in use" in proportion to increasing the amount of the oily ingredient cannot be observed. In addition, in view of obtaining a substantial feeling in use without stickiness, it is particularly preferable that the oily ingredient be added in a range of 1% by weight or more and 10% by weight or less based on the total weight of the composition.

[0022] In the emulsified composition of the present invention, it is characterized in that the number-average particle size (diameter) of the oily ingredient of the inner phase formed as a result of emulsification (in the following, also referred to as "emulsified particles") is in a range of 1  $\mu\text{m}$  or less. Means for making the number-average particle size of the oily ingredient of the "inner phase" which corresponds to a liquid part in the form of a microparticle dispersed in the emulsified composition of the present invention be 1  $\mu\text{m}$  or less is not particularly restricted. Therefore, any means is acceptable so long as the microemulsification can be carried out.

[0023] Commonly, when an emulsion is formed by emulsifying an oily ingredient and an aqueous ingredient, the desirable emulsified particles having the number-average particle size of 1  $\mu\text{m}$  or less can be prepared according to an interface scientific emulsification method including, for example, a nonaqueous emulsification method (Takashi OHASHI: Japanese Examined Patent Application, Second Publication No. Sho 57-29213), a D-phase emulsification method (H. Sagitani: J. Am. Oil Chem. Soc., 58, 738 (1981)), or a phase inversion temperature emulsification method (K. Shinoda: J. Colloid

Interface Sci., 24, 4 (1969), and the like)), by means of various emulsifying devices which are commonly used, such as a homomixer, a homodisper, a homogenizer, a colloid mill, a pebble mill, or an ultrasonic emulsifier, and the like. However, in the case of preparing the emulsified particles by the means described above, it is not necessarily advantageous that there be some restrictions on the preparation regarding an amount of a humectant, the selection of a surfactant, and the like.

[0024] It is advantageous to use, as means for preparing the emulsified composition of the present invention, a "high-pressure emulsifier" carrying out emulsification by injecting a liquid from a nozzle under a high pressure ranging from 100 to 1000 bar, since the high-pressure emulsifier does not have the restrictions on the preparation of emulsions as described above which the commonly used emulsifiers have. In detail, as examples thereof, mention may be made of commercially available products such as a Microfluidizer, a Nanomizer, or a Mantongourin (all trade names). Among these commercially available products, the Nanomizer corresponds to a device in which the emulsified microparticles can be obtained by collision between an oily ingredient and an aqueous ingredient under high pressure from nozzles having a connected groove. The particle size of the emulsified particles commonly prepared by the emulsifier is very small and fine such as in a range of 1  $\mu\text{m}$  or less. For this reason, it is a particularly preferable emulsifier as the emulsification means when the emulsified compositions of the present invention are prepared.

[0025] The emulsified compositions of the present invention comprising the essential components described above and having the particle size of the emulsified particles within the range

described above exhibit superior features that a substantial feeling in use is imparted upon application to the skin, stability at low temperature is superior, and high safety is afforded.

[0026] The emulsified compositions of the present invention can be formulated into cosmetics basically in the form of an oil-in-water type (O/W type) emulsion. In detail, as examples of the formulations and forms, mention may be made of a milk, a cream, a cleansing cream, an emulsified foundation, and the like.

[0027] To the emulsified compositions of the present invention, in addition to the essential components described above, various agents and/or ingredients of a base agent, and the like which are commonly used in the compositions for use in cosmetics can be added according to the effects to be imparted or formulations described above, unless the desirable effects of the present invention are impaired. In the following, these agents and base agent ingredients are exemplified. However, it should be understood that the agents and the ingredients of a base agent, and the like which can be added to the emulsified compositions of the present invention are not restricted to these examples.

[0028] Humectants, examples of which include, for example, polyethylene glycol, propylene glycol, dipropylene glycol, 1,3-butylene glycol, hexylene glycol, glycerol, diglycerol, xylitol, multitol, maltose, D-mannitol, sorbitol, and the like, can be added to the emulsified compositions of the present invention.

[0029] In addition, ultraviolet absorbents, examples of which include, for example, a benzoic acid type UV absorbent such as para-aminobenzoic acid; an anthranilic acid type UV absorbent such as methyl anthranilate; a salicylic acid type UV absorbent such as octyl salicylate, or phenyl salicylate; a cinnamic acid type UV absorbent such as isopropyl para-methoxycinnamate, octyl para-methoxycinnamate, or glyceryl di-para-methoxycinnamate mono-2-ethylhexanoate; a benzophenone type UV absorbent such as 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, or 2-hydroxy-4-methoxybenzophenone-5-sulfonic acid; urocanic acid, 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, 4-tert-butyl-4'-methoxybenzoyl methane, and the like can be added to the emulsified compositions of the present invention.

[0030] In addition, as an antibacterial agent, benzoic acid, salicylic acid, carbolic acid, sorbic acid, an ester of para-oxybenzoic acid, para-chloro-metacresol, chlorohexidine chloride, trichlorocarbanide, phenoxyethanol, or the like can be added to the emulsified compositions of the present invention.

[0031] In addition, as common agents, vitamins such as vitamin A oil, retinol, retinol palmitate, inositol, pyridoxine hydrochloride, benzyl nicotinate, nicotinamide, DL- $\alpha$ -tocopherol nicotinate, magnesium phosphate ascorbate, vitamin D2 (ergocalciferol), dl- $\alpha$ -tocopherol, dl- $\alpha$ -tocopherol acetate, pantothenic acid, biotin, and the like; hormones such as estradiol, ethynyl estradiol, and the like; amino acids such as arginine, aspartic acid, cystine, cysteine, methionine, serine, leucine, tryptophan, and the like; anti-inflammation agents such as allantoin, azulene, and the like; whitening agents such as arbutin and the like; astringents such as zinc chloride,



tannic acid, and the like; cooling agents such as L-menthol, camphor, and the like; sulfur, lysozyme chloride, pyridoxine hydrochloride,  $\gamma$ -oryzanol, and the like can be added to the emulsified compositions of the present invention.

[0032] In addition, as various extracts, *Houttuynia cordata* extract, obaku extract, melilote extract, white dead nettle extract, glycyrrhiza extract, peony extract, soapwort extract, luffa extract, quina extract, strawberry geranium extract, *Sophora angustifolia* extract, candock extract, anise extract, primrose extract, rose extract, *Rehmannia glutinosa* extract, lemon extract, *Lithospermum erythrorhizon* extract, aloe extract, sweetflag root extract, eucalyptus extract, field horsetail extract, sage extract, thyme extract, green tea extract, marine algae extract, cucumber extract, clove extract, raspberry extract, melissa extract, ginseng extract, horse chestnut extract, peach extract, peach leaf extract, mulberry extract, cornflower extract, witch-hazel leaf extract, placenta extract, thymus gland extract, silk extract, and the like can be added to the emulsified compositions of the present invention.

[0033] Furthermore, among ingredients of a base agent which can be added to the emulsified compositions of the present invention, as liquid fats and oils, linseed oil, camellia oil, macadamia nut oil, corn oil, mink oil, olive oil, avocado oil, sasanqua oil, castor oil, safflower oil, jojoba oil, sunflower oil, almond oil, rapeseed oil, sesame oil, soybean oil, peanut oil, triglycerol, glycerol trioctanoate, glycerol triisopalmitate, and the like can be added to the emulsified compositions of the present invention.

[0034] In addition, as ester oils, cetyl octanoate, hexyl laurate, isopropyl myristate, octyl palmitate, isocetyl stearate, isopropyl isostearate, octyl isopalmitate, isodecyl oleate, glyceryl tri 2-ethylhexanoate, pentaerythrityl tetra 2-ethylhexanoate, 2-ethylhexyl succinate, diethyl sebacate, and the like can be added to the emulsified compositions of the present invention.

[0035] In addition, as hydrocarbon oils, liquid paraffin, squalane, squalene, paraffin, isoparaffin, ceresin, and the like can be added to the emulsified compositions of the present invention.

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[0036] In addition, as silicone oils, a chain silicone oil such as dimethyl polysiloxane, methylphenyl polysiloxane, methylhydrogen polysiloxane; a cyclic silicone oil such as octamethyl cyclotetrasiloxane, decamethyl cyclopentasiloxane, dodecamethyl cyclohexasiloxane, and the like can be added to the emulsified compositions of the present invention.

[0037] In addition, as lower alcohols, methanol, ethanol, propanol, isopropanol, and the like can be added to the emulsified compositions of the present invention.

[0038] In addition, as water-soluble polymers, plant type polymers such as arabic gum, carageenane, pectin, agar, quince seed (marmelo), starch, algae colloid (brown algae extract), and the like; microorganism type polymers such as dextran, pullulan, and the like; animal polymers such as collagen, casein, gelatin, and the like; starch type polymers such as carboxymethyl starch, methylhydroxypropyl starch, and the like; cellulose type polymers such as methyl cellulose, nitro cellulose, ethyl

cellulose, methylhydroxypropyl cellulose, hydroxyethyl cellulose, and the like; alginic acid type polymers such as sodium alginate, and the like; vinyl type polymers such as carboxyvinyl polymer (CARBOPOL and the like); polyoxyethylene type polymers; polyoxyethylene polyoxypropylene copolymer type polymers; acryl type polymers such as sodium polyacrylate, polyacrylamide, and the like; inorganic type water-soluble polymers such as bentonite, magnesium aluminum silicate, laponite, and the like; and the like can be added to the emulsified compositions of the present invention.

[0039] In addition, as sequestering agents, sodium edetate, sodium metaphosphate, and the like can be added to the emulsified compositions of the present invention.

[0040] In addition, as neutralizers, 2-amino-2-methyl-1-propanol, potassium hydroxide, sodium hydroxide, triethanolamine, sodium carbonate, and the like can be added to the emulsified compositions of the present invention.

[0041] In addition, as antioxidants, ascorbic acid,  $\alpha$ -tocopherol, dibutylhydroxyanisole, and the like can be added to the emulsified compositions of the present invention.

[0042] The agents and the ingredients of a base agent described above can be used in a free state, or if they are able to form salts, they can be used in the form of a salt of an acid or a base, or alternatively, if they have carboxylic acid groups, they can be used in the form of an ester. Furthermore, appropriate fragrances, pigments, and the like can be added to the emulsified compositions of the present invention, as necessary, within a range which does not impair the desired

effects of the present invention. The formulations of the present invention will be explained in detail in Examples described below.

[0043]

[Examples] In the following, the present invention will be explained in detail by reference to Examples and the like. However, it should be understood that the technical scope of the present invention is not restricted to these Examples. In the following, "% by weight" means percent by weight with respect to total weight of the composition, unless otherwise indicated.

[0044] First, evaluation methods of the emulsified compositions of the present invention in the Examples will be explained.

<Sensory test by female specialist panelists>

The sensory test with regard to a feeling in use (a substantial feeling in use) was carried out by 10 female specialist panelists.

Evaluation criteria regarding a "substantial feeling in use"  
◎: superior (9 or more panelists among 10 panelists evaluate it as being good)

○: good (7 or 8 panelists among 10 panelists evaluate it as being good)

△: fair (3 or more and 6 or less of panelists among 10 panelists evaluate it as being good)

×: inferior (2 or less panelists among 10 panelists evaluate it as being good)

[0045] <Test 1 of stability at low temperature>

The emulsified compositions were allowed to stand in a thermostatic chamber at 0°C, and the state of the same after 6 months was observed by means of a polarization microscope.

Evaluation criteria of stability at low temperature

○: Presence of crystals was not observed at all.

△: Presence of crystals was slightly observed.

×: Presence of crystals was clearly observed.

[0046] <Method for measuring the number-average particle size of the emulsified particles>

The photograph of the emulsion immediately after preparation was taken by means of an optical microscope photography, and was subsequently subjected to visual observation.

[0047] [Examples 1 and 2 and Comparative Examples 1 to 3]  
The emulsified compositions having the preparations shown in Table 1 were prepared. Subsequently, "substantial feeling in use" and stability at low temperature of the same were evaluated and the number-average particle size of the emulsified particles was measured. The results of the evaluation and the like are also shown in Table 1.

[0048]

Table 1

| Ingredients          | Example 1 | Example 2 | Comparative Example 1 | Comparative Example 2 | Comparative Example 3 |
|----------------------|-----------|-----------|-----------------------|-----------------------|-----------------------|
| A. Oil phase         |           |           |                       |                       |                       |
| Liquid paraffin      | 15.0      | 15.0      | 15.0                  | 15.0                  | 15.0                  |
| Dimethylpolysiloxane | 1.0       | 1.0       | 1.0                   | 1.0                   | 1.0                   |
| Stearyl alcohol      | 4.0       | 4.0       | 4.0                   | -                     | -                     |
| Stearic acid         | 1.0       | -         | 1.0                   | -                     | -                     |

|   |                              |                              |                   |                              |                   |
|---|------------------------------|------------------------------|-------------------|------------------------------|-------------------|
| B. Aqueous phase  |                              |                              |                   |                              |                   |
| PEMULEN TR-1  | 0.2                          | 0.2                          | 0.2               | 0.2                          | 0.2               |
| CARBOPOL 941  | 0.1                          | 0.1                          | 0.1               | 0.1                          | 0.1               |
| Potassium hydroxide   | 0.1                          | 0.1                          | 0.1               | 0.1                          | 0.1               |
| Glycerol  | 10.0                         | 10.0                         | 10.0              | 10.0                         | 10.0              |
| Ethyl alcohol   | 1.0                          | 1.0                          | 1.0               | 1.0                          | 1.0               |
| Methyl paraoxybenzoate  | 0.5                          | 0.5                          | 0.5               | 0.5                          | 0.5               |
| Ion-exchanged water   | balance                      | balance                      | balance           | balance                      | balance           |
| Preparation method  | High-pressure emulsification | High-pressure emulsification | HM emulsification | High-pressure emulsification | HM emulsification |
| Substantial feeling in use  | ◎                            | ○                            | ◎                 | ×                            | ×                 |
| Stability at low temperature  | ○                            | ○                            | ×                 | ○                            | ○                 |
| Number-average particle size of the emulsified particles ( $\mu$ m) | 1 $\mu$ m or less            | 1 $\mu$ m or less            | 5 $\mu$ m         | 1 $\mu$ m or less            | 5 $\mu$ m         |

[0049] <Preparation method>

The oily ingredients were heated to be in the form of a liquid. The oily liquid was added to the aqueous phase ingredients. The mixture was emulsified by means of a rotary type emulsifier (homomixer: HM) or a high-pressure emulsifier (nanomizer).

[0050] In both Example 1 and Example 2, good substantial feeling in use and good stability at low temperature were together exhibited. In Example 1, stearic acid corresponding to one of the suitable fatty acids to be added in the present invention was added to the preparation of Example 2. For this reason, the superior "substantial feeling in use" was particularly exhibited.

[0051] In addition, in Comparative Example 1 in which the particle size of the emulsified particles was 5  $\mu$  m even if the ingredients of Comparative Example 1 were the same as those

of Example 1, stability at low temperature was inferior although the substantial feeling in use was equivalent to that of Example 1. From these results, it was shown that in order to impart stability at low temperature, it is necessary to adjust the particle size of the emulsified particles to 1  $\mu\text{m}$  or less.

[0052] Furthermore, in Comparative Example 2 in which an "oily ingredient in a solid state at room temperature" was not included, the "substantial feeling in use" was inferior, although superior stability at low temperature was exhibited. From these results, it was shown that adding an "oily ingredient in a solid state at room temperature" was necessary to impart a "substantial feeling in use" to the emulsified compositions of the invention. Next, the stability at low temperature of the composition according to Example 1 was further tested.

[0053] <Test 2 of stability at low temperature>

Measurement of X-ray diffraction

Measurements of X-ray diffraction were carried out in the composition of Example 1 and the comparative composition of Comparative Example 1. According to the X-ray diffraction, from the results, the degree of proceeding crystallization of the fatty acid can be mainly determined. Fig. 1 is a graph showing the result of X-ray diffraction of the emulsified composition according to Example 1 stored for 6 months at 0°C. Fig. 2 is a graph showing the result of X-ray diffraction of the comparative emulsified composition according to Comparative Example 1 stored for 6 months at 0°C.

[0054] It can be seen that in Comparative Example 1, there is a diffraction line of the short surface side of the fatty acid crystal in the vicinity of 15 to 25 of the angle ( $2\theta$ ),

and for this reason, crystallization (unstabilization) proceeds. On the other hand, it can be seen that in Example 1, a diffraction line of the short surface side of the fatty acid crystal is hardly observed, and for this reason, crystallization proceeds at a low rate.

[0055] In the following, formulations of various emulsified compositions according to the present invention are described (the preparation method of each of the compositions was carried out according to the same manner as described in Example 1 and Example 2 described above). In addition, the tests described above were carried out in these emulsified compositions. As a result, all of the emulsified compositions exhibited a "substantial feeling in use" and had superior stability at low temperature.

[0056] [Example 3]

|                               | % by weight |
|-------------------------------|-------------|
| A. Oily phase                 |             |
| Dimethylpolysiloxane          | 5.0         |
| Isopropyl myristate           | 3.0         |
| Liquid paraffin               | 2.0         |
| $\alpha$ -tocopherol          | 0.1         |
| Behenic acid                  | 9.0         |
| B. Aqueous phase              |             |
| Triethanolamine               | 0.35        |
| Propylene glycol              | 15.0        |
| Methyl paraoxybenzoate        | 0.2         |
| PEMULEN TR-2                  | 1.0         |
| Hydroxypropyl methylcellulose | 0.5         |
| Ion-exchanged water           | remainder   |



[0057] [Example 4]

% by weight

## A. Oily phase

|                                  |      |
|----------------------------------|------|
| Jojoba oil                       | 3.0  |
| dl- $\alpha$ -tocopherol acetate | 0.1  |
| Butyl paraoxybenzoate            | 0.1  |
| Behenyl alcohol                  | 0.5  |
| Microcrystalline wax             | 0.1  |
| Decamethylcyclopentasiloxane     | 20.0 |

## B. Aqueous phase

|                     |           |
|---------------------|-----------|
| Dipropylene glycol  | 7.0       |
| Trisodium edetate   | 0.05      |
| PEMULEN TR-1        | 3.0       |
| Sodium hydroxide    | 1.0       |
| Ethanol             | 9.0       |
| Ion-exchanged water | remainder |

[0058] [Example 5]

% by weight

## A. Oily phase

|                          |      |
|--------------------------|------|
| Macadamia nut oil        | 10.0 |
| Methylphenylpolysiloxane | 2.0  |
| Diethyl sebacate         | 4.0  |
| Vitamin A oil            | 0.01 |
| Beeswax                  | 5.0  |
| Vaseline                 | 12.0 |

## B. Aqueous phase

|                               |      |
|-------------------------------|------|
| 1,3-butylene glycol           | 10.0 |
| Propylene glycol              | 5.0  |
| Albutine                      | 5.0  |
| Magnesium ascorbate phosphate | 1.0  |
| Placenta extract              | 0.05 |

|                       |           |
|-----------------------|-----------|
| Luffa extract         | 0.5       |
| PEMULEN TR-1          | 0.02      |
| CARBOPOL 941          | 0.1       |
| Ethyl paraoxybenzoate | 0.3       |
| Phenoxyethanol        | 0.3       |
| Ion-exchanged water   | remainder |

[0059]      [Example 6]

% by weight

A. Oily phase

|                            |      |
|----------------------------|------|
| Squalane                   | 10.0 |
| Diocetyl succinate         | 5.0  |
| Octyl paramethoxycinnamate | 0.1  |
| Cetostearyl alcohol        | 0.5  |
| Retinol palmitate          | 0.4  |

B. Aqueous phase

|                        |           |
|------------------------|-----------|
| PEMULEN TR-1           | 0.01      |
| PEMULEN TR-2           | 0.05      |
| Sorbitol               | 5.0       |
| Ascorbic acid          | 1.0       |
| Methyl paraoxybenzoate | 0.5       |
| Ion-exchanged water    | remainder |

[0060]

[Effects of the Invention] According to the present invention, in emulsified compositions, which do not need to use a surfactant as emulsifying means in order to ensure high safety, an emulsified composition sufficiently maintaining "a substantial feeling in use", and exhibiting superior stability at low temperature can be provided.

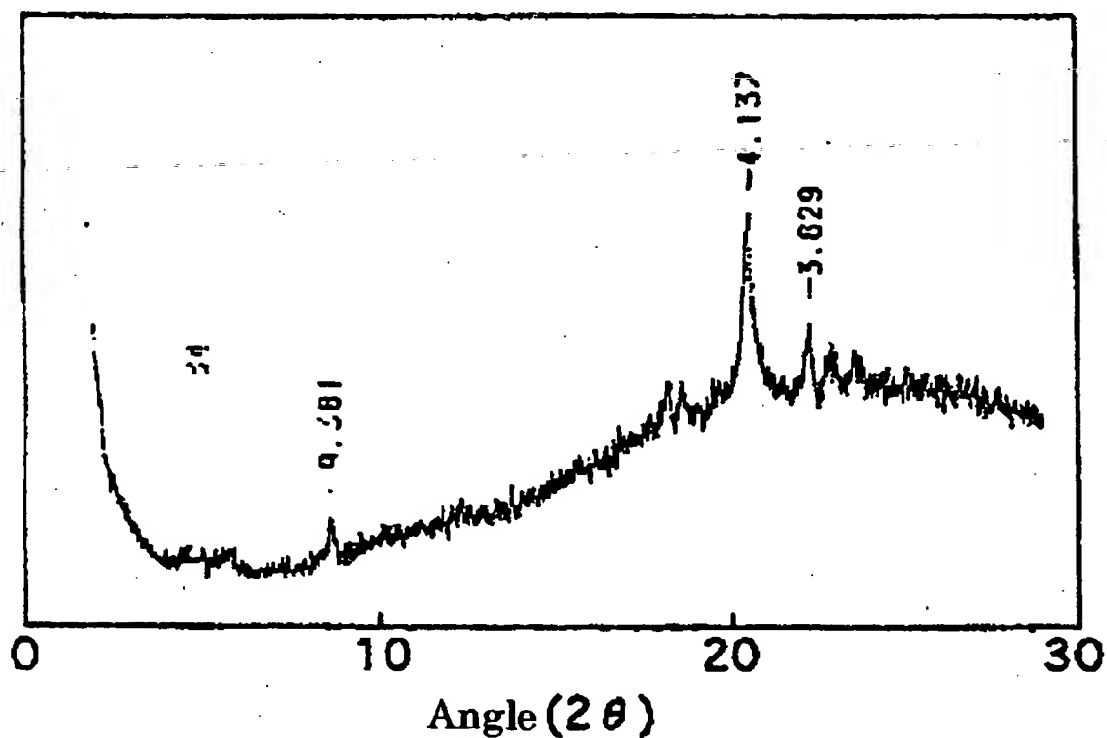
[Brief Explanation of the Drawings]

[Fig. 1] Fig. 1 is a drawing showing the results of X-ray diffraction of the emulsified composition of the present invention according to Example 1.

[Fig. 2] Fig. 2 is a drawing showing the results of X-ray diffraction of the comparative emulsified composition according to Comparative Example 1.

[Fig. 1]

Fig. 1



[Fig. 2]

Fig. 2

